

9. Groundwater Dating

Water moves through the hydrological cycle at different rates. Water vapor has a mean residence time in the atmosphere of only about 2 weeks. Runoff in first order streams and high elevation catchments are highly responsive to precipitation and melting, with circulation rates measured in hours to days. Large rivers draining major sectors of the continents integrate precipitation on the scale of weeks to months, and the mean residence time for precipitation in large lakes is on the order of decades to over one hundred years.

Groundwater circulates on quite a different time scale. Alluvial aquifers along drainage channels or shallow bedrock aquifers can respond rapidly to recharge and have groundwaters with mean residence times on the order of months to years. Deeper and confined aquifers host groundwaters that may circulate over hundreds to thousands of years. Others still may retain groundwaters recharged under climate conditions that no longer exist, and so are “fossil” groundwaters, no longer linked to active meteorological processes.

Surface water flow is extensively monitored by measurement of discharge rates at critical points in the drainage network, as input for flood control, hydroelectric generation and water resources management. Although groundwater resources are several times greater than all surface waters, their slow movement and cryptic nature precludes direct measurement of natural flow. Groundwater flow can be determined from hydraulic conductivity and gradient distributions. It can also be measured from geochemical means.

Determining the mean subsurface residence time or “age” of groundwater is important for several reasons. Groundwater age translates roughly to renewability, which is a critical factor for water resource planning. Groundwater resources identified as being modern are regularly recharged and can be sustainably developed, whereas exploiting thousands of years old groundwaters may be mining the resource. The age of a groundwater relates also to its vulnerability to contamination. Modern groundwaters will have more direct pathways from the surface, facilitating infiltration of contaminants from surface activities in the recharge area. Old groundwaters tend to be relatively isolated from the surface, with a greater potential for attenuation of contamination.

Groundwater Mean Residence Time and “Age”

Groundwater recharge occurs in most cases by infiltration of precipitation and runoff through soils, alluvial sediments or bedrock outcrop into the subsurface. It may move slowly through an unsaturated zone, or directly to the water table. In any case, some of the water will take a longer pathway through the pores and fissures, some will take a more direct route. This process of hydrodynamic dispersion mixes waters having recharged at different times. The difference in time may only be hours or days for soil waters in the first several centimeters. In the unsaturated zone and just below the water table, a given volume may integrate groundwaters recharged over a span of weeks to months. Deeper in the aquifer, the groundwater will include components recharge over a greater range of months to years. At each point, the groundwater will have a mean age, and a range of ages.

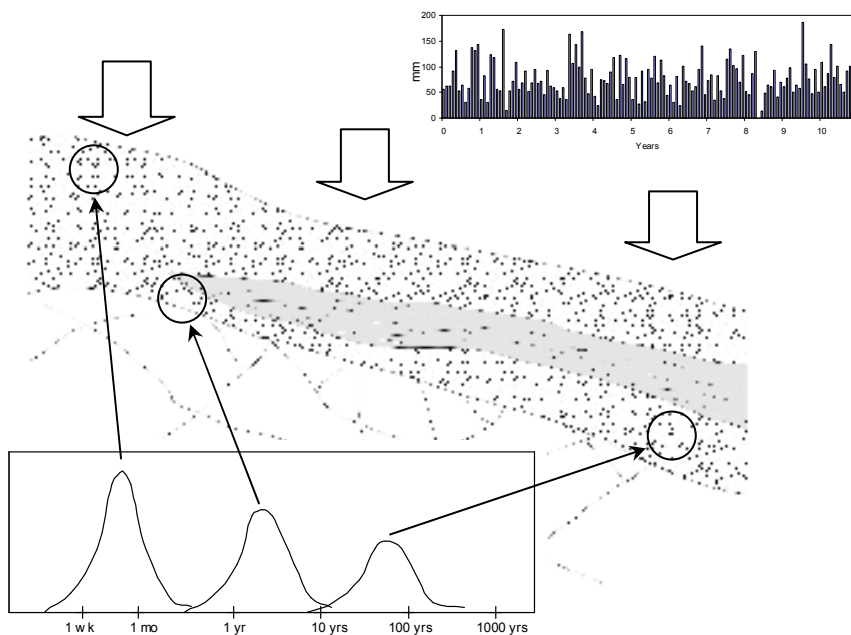


Fig. 9-1 Ranges of ages for groundwaters at different points along flow paths. Input precipitation represents a unique time series, which is transformed in the subsurface by hydrodynamic mixing. Vertical scale on curves represent relative age distribution for groundwater at that point. Peak represents the mean age of groundwater.

Classifying the age of groundwater tends to be method driven. Tritium allows identification of water recharged within the past 50 years, and such waters

are termed “modern”. With the complications of carbon sources and sinks in the subsurface, the resolution of radiocarbon allows identification of “old” groundwaters recharged more than one thousand to about perhaps twenty thousand years before present. Groundwaters with little to no radiocarbon remaining are termed “ancient” or “fossil”, and are often no longer linked to modern meteorological processes. Groundwaters falling within the gap between these two methods, i.e. tritium-free groundwaters with no evidence of radiocarbon decay, are termed “submodern”.

Despite advances with other geochemical and isotopic tools, tritium and radiocarbon remain the most accessible and reliable isotopic methods for dating groundwaters. New and interesting radioisotopes are discussed in a subsequent section.

Tritium and “Modern” Groundwaters

Tracers of modern meteoric water should provide either a characteristic signal that can be preserved in recently recharged waters, or an input that decays once removed from the meteoric source. Tritium (^3H or T) has both. It is a short-lived radioisotope that is naturally produced in the atmosphere and directly incorporated into precipitation. It also has a characteristic signal from atmospheric tests of nuclear weapons in the 1960s. Both decay and position of the thermonuclear peak can provide groundwater age information.

Tritium is a short-lived isotope of hydrogen with a half-life of 12.32 years (Lucas and Unterweger, 2000). It is directly incorporated into the water molecule (H^3HO or HTO) and so is the only radioisotope that actually dates groundwater.

Unlike CFCs, ^{85}Kr and other modern tracers which require very strict sampling protocols, contamination by tritium in atmospheric water vapor during sampling is not a concern. Measurement by β^- counting directly on water or on electrolytically-enriched water is routine and inexpensive, and provides unambiguous evidence for modern recharge.

Atmospheric testing of thermonuclear weapons from May 1951 to 1976 generated significant reservoirs of anthropogenic tritium in the upper atmosphere that has only recently been washed out to natural background levels. Its decay from natural, pre-bomb levels is such that it cannot normally be detected in groundwaters recharged before about 1950. Modern

groundwaters are then younger than about 45 years relative to the mid-1990s. Tritium-free groundwaters are considered “submodern” or older.

Tritium concentration units

The use of tritium in dating groundwaters relies on knowledge of its concentration in meteoric waters, to which both natural and anthropogenic sources contribute. The most widely used unit of concentration of tritium is the Tritium Unit, or TU, where:

$$\begin{aligned} 1 \text{ TU} &= 1 \text{ T atom per } 10^{18} \text{ H atoms} \\ &= 6.7 \cdot 10^7 \text{ T atoms per kg H}_2\text{O} \\ &= 1.1 \cdot 10^{-16} \text{ mol/kg H}_2\text{O} \\ &= 3.33 \cdot 10^{-10} \text{ ppb (gfw for T is 3)} \end{aligned}$$

Enrichment methods can detect to below 1 TU, which makes this measurement one of the most sensitive in geochemistry. Measurement is done by scintillation counting of β^- decay events, or by atom-counting of ^3He in sealed water samples that were pumped of gases and stored for a given period of decay.

The question of the radiological hazard of tritium often comes up, which requires expressing tritium concentrations in units of radioactivity. Radioactivity is expressed in terms of decay events per time. The original unit used is the curie, Ci, which is equal to the radioactivity of 1 g of ^{226}Ra – the isotope of radium that Curie isolated from the ^{238}U decay series. The bequerelle, Bq, is now used, defined as one nuclear disintegration per second ($1 \text{ Ci} = 3.7 \cdot 10^{10} \text{ Bq}$)

The radioactivity of a radionuclide is equal to its change in concentration with time, $\partial N / \partial t$. Converting between units of concentration (atoms per volume or mass) and radioactivity (decay events per time) is done with the decay constant, λ , which is based on the half-life of the radionuclide:

$$\begin{aligned} \lambda &= \frac{\ln 2}{t_{1/2}} \text{ (time}^{-1}\text{)} && \text{decay constant} \\ -\frac{\partial N}{\partial t} &= \lambda N \text{ (decay / time)} && \text{radioactive decay} \end{aligned}$$

Radionuclides with short half-lives like tritium are then much more radioactive than one with long half-life (e.g. common uranium, ^{238}U , $t_{1/2} = 4.5 \cdot 10^9 \text{ y}$) for an equivalent concentration or mass.

For tritium decay, radioactive decay is the rate of change in tritium concentration is a function of its concentration, N (T atoms per liter). The decay constant, λ , for tritium is:

$$\lambda = \frac{\ln 2}{t_{1/2}} = \frac{0.693}{12.32} = 0.05626 \text{ yr}^{-1}$$

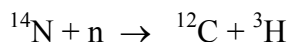
The radioactivity is simply the product of its concentration and decay constant. The radioactivity of 1 L water with 1 TU ($N = 6.67 \cdot 10^7$ atoms T, from above) would then yield:

$$\begin{aligned} \text{radioactivity} &= \lambda N \\ &= 0.05626 \cdot 6.67 \cdot 10^7 \text{ disintegrations per year per liter} \\ &= 3.76 \cdot 10^6 \text{ disintegrations per year per liter, or} \\ &= 0.119 \text{ disintegrations per second per liter} \\ &= 0.119 \text{ Bq/L (Bq = 1 disintegration per second)} \end{aligned}$$

The radioactivity of water with natural levels of tritium is low, and well below the WHO recommended limit of 7800 Bq/L. For water to exceed this limit, it would need over 65,000 TU, a level never recorded in precipitation or natural groundwaters. Even at the peak, anthropogenic tritium fallout was not a radiological threat to health. Minor amounts of ^3H are presently released to the atmosphere by nuclear power plants, nuclear fuel reprocessing facilities, preparation of weapons-grade nuclear material and by the manufacturers of tritiated paints used in illuminating dials and tritium gas (T_2) for self-illuminating emergency signs.

Tritium production and decay

Natural tritium is formed in the stratosphere where high-energy cosmic radiation produces a secondary neutron flux. Tritium is produced by collisions of these neutrons with N_2 , following the reaction:



Tritium decays with a half-life of 12.32 years to ^3He by release of a beta particle:



Chemically, the newly produced tritium still behaves like stable hydrogen, ^1H , and so combines with atmospheric oxygen or hydroxide to form a water

molecule. Most cosmogenic tritium is generated in the stratosphere, and enters the troposphere through mixing in the spring.

The natural neutron activation process is emulated in nuclear reactors and in thermonuclear bombs, making anthropogenic activities major sources of environmental tritium. This was first predicted and observed in the 1950s during tests of hydrogen fusion bombs where the release of a huge neutron flux activates ^{14}N in the atmosphere. A decade of American and Soviet atmospheric tests of thermonuclear weapons in the upper atmosphere began on November 1 1952 with the detonation of the 10.4 megaton device, Mike, and ended in 1962 with a final spectacular release of over 140 megatons of fusion energy. The fireball that Mike generated was over 3 miles wide, with a neutron flux calculated to be greater than that of a supernova (rate of neutrons released, not total neutrons, of course). The consequence was a dramatic increase of tritium in precipitation, reaching a peak of over 5000 TU in the northern hemisphere (Fig. 9-2).

The final year of megaton tests (1962) generated a huge peak, which appeared in precipitation in the spring of 1963. This 1963 peak became a marker used in many hydrological studies. Concentrations of ^3H in precipitation have now stabilized at close to pre-bomb, cosmogenic levels.

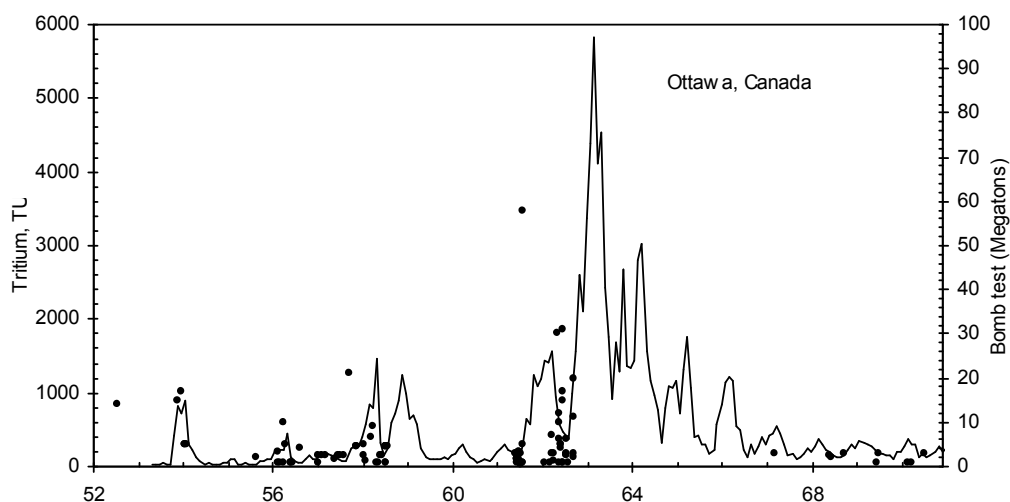


Fig. 9-2 The production of anthropogenic tritium as a consequence of above-ground testing of thermonuclear (hydrogen-fusion) weapons. The Ottawa record is the longest continuously-monitored tritium record for precipitation, begun in 1951 by Dr. Bob Brown, a radiochemist with Atomic Energy of Canada Limited, Chalk River, Ontario, and maintained by the Environmental Isotope Laboratory, University of Waterloo, Ontario. Available through IAEA/WMO (2001). The limited test ban treaty of 1963 between the USSR and USA was followed by a sharp decline in atmospheric ^3H , reflecting mixing with ocean water. Annual “leaks” of tritium from the stratosphere to the troposphere occur each spring with the northward shift of the Hadley cells and the jet stream.

The Soviet tests were conducted in eastern Kazakhstan at about 50°N, and the American tests in the Marshall Islands of the eastern Pacific Ocean at 12°N. Levels in the southern hemisphere were much lower and lagged northern hemisphere precipitation by a few years. Further tests by France and China sustained levels briefly in the 1970s. Through mixing with the oceans, atmospheric levels declined rapidly, essentially reaching background levels by the 1990s.

Although fission from detonation of uranium and plutonium bombs began on July 16, 1945, the neutron flux from these comparatively small and low-level fission tests, had no impact on global meteoric ^3H .

The asymmetric production of thermonuclear tritium between the two hemispheres was manifested by both the timing and magnitude of the 1963 tritium peak (Fig. 9-3). While the tritium peak exceeded 5000 TU as far north as Ottawa (at the latitude of the Soviet tests), in the southern hemisphere it only attained a few hundred tritium units.

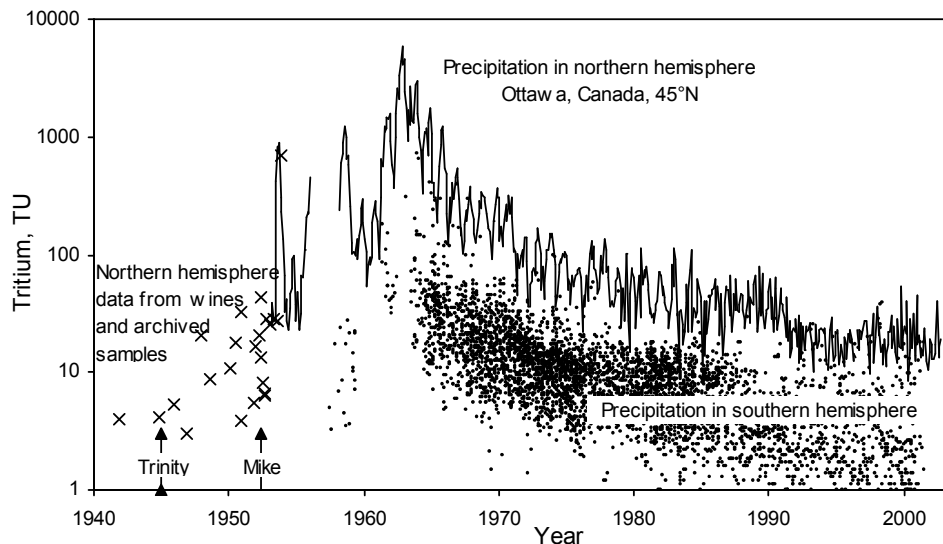
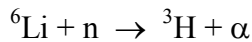


Fig. 9-3 Latitudinal variations in magnitude of the 1963 thermonuclear tritium peak. The high production from Soviet tests in Kazakhstan by the American tests in the Marshall Islands is seen in the high fallout for the northern latitudes (solid line – Ottawa, Canada). Transfers to the southern hemisphere (dots) are attenuated by a factor of about 10. Trinity was the earliest fission bomb test. The first hydrogen fusion bomb test was code-named Mike.

The decline of tritium in precipitation is not due only to decay, which decreases tritium levels by only 5.5% per year. Radioactive decay from

example initial values follows straight lines on the log-normal plot in Fig. 9-4. Attenuation by the oceans and groundwaters has reduced levels considerably faster than by decay alone.

Unlike other radionuclides like ^{129}I and ^{36}Cl , subsurface production of tritium is generally negligible, but can be measured. A neutron flux is present in the subsurface from spontaneous fission of U. This can result in ^3H production through the fission of ^6Li according to:



However, tritium's short half-life allows insufficient time for significant accumulation. Groundwater in U and Li rich rocks seldom have over 0.5 TU, although in uranium ore zones levels may be substantially higher. In most aquifers, geogenic ^3H is insignificant.

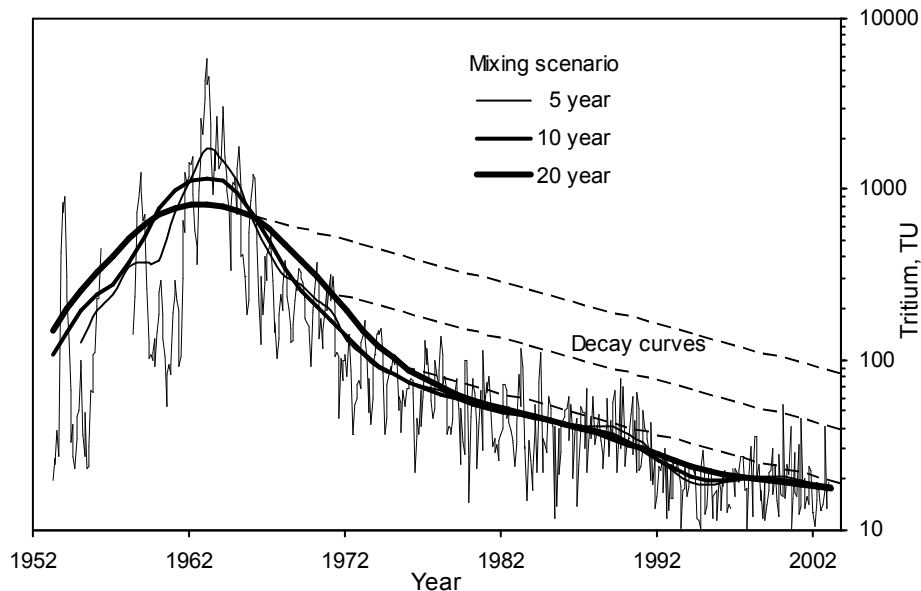


Fig. 9-4 Tritium concentrations in precipitation from Ottawa, Canada. Composite monthly samples plotted in light continuous line. Multi-year mixing scenarios are shown for 5, 10 and 20 year cases, using a normal distribution function to weight the annual tritium contributions.

Global distribution of natural tritium

Hydrogeologists are entering a new era for the use of tritium in groundwater dating. The thermonuclear peak of 1963 has been largely flushed from most actively recharged aquifers, and precipitation is essentially back to pre-anthropogenic levels controlled by natural production. This replaces the variable input function from the bomb testing era with a relatively constant input function. In the next section, it is shown how this facilitates the use of the decay equation to determine groundwater age.

Although tritium in precipitation is back to natural levels, there are still spatial variations and seasonal variations. The following maps present background levels for different regions.

Dating Groundwaters with Tritium

The presence of tritium in groundwater, at its most fundamental level of interpretation, is an indication that at least some component of modern recharge. Modern recharge is taken to be younger than the onset of thermonuclear bomb tests. Today, this gives a time constraint of some 50 years. However, different scenarios for groundwater flow and discharge will greatly affect the concentration of tritium, and so hydrogeological constraints should be taken into consideration. Groundwater mixing, particularly at the discharge zone in spring vents or in open boreholes, greatly affects the range of groundwater ages. With the tritium input function from the past 50 years that varies over several orders of magnitude (Fig. 9-4), the relative contributions from recent and pre-1950s precipitation will greatly affect the measured tritium in groundwater.

More refined interpretations are based on the level of tritium, rather than simply a measurable concentration. At this point, some knowledge of the history of tritium in local precipitation is required. The following points should be considered:

- In most regions, tritium concentrations in precipitation are now back to natural production levels.
- Bomb tritium, found in groundwaters recharged prior to about 1990, is higher in the mid latitude northern hemisphere than in low latitudes, tropics and southern hemisphere.

- Coastal precipitation generally has much lower tritium concentrations than continental (interior) stations. This is true for precipitation during the thermonuclear era as well as today and is due to the lower degree of mixing with upper atmosphere vapor experienced by coastal precipitation systems.
- Groundwaters with low tritium levels (close to 1 TU) are often mixtures of modern groundwaters and submodern groundwaters (greater than at least 50 years old).

Groundwater age from the movement of the 1963 ^3H peak: The use of tritium to determine groundwater age is greatly aided by sampling sequentially over a period of time. Temporal changes in tritium may relate to changing source term (i.e. the declining bomb input) or changes in mixing of different groundwaters. In the ideal case, time-series measurements with rising tritium levels followed by declining levels show the movement of the 1963 peak, and from this groundwater flow velocity can be calculated. Similarly, spatial variations can be used to identify the 1963 peak. The groundwaters in Fig. 9-5 demonstrate this, and allow calculation of recharge rates at this site.

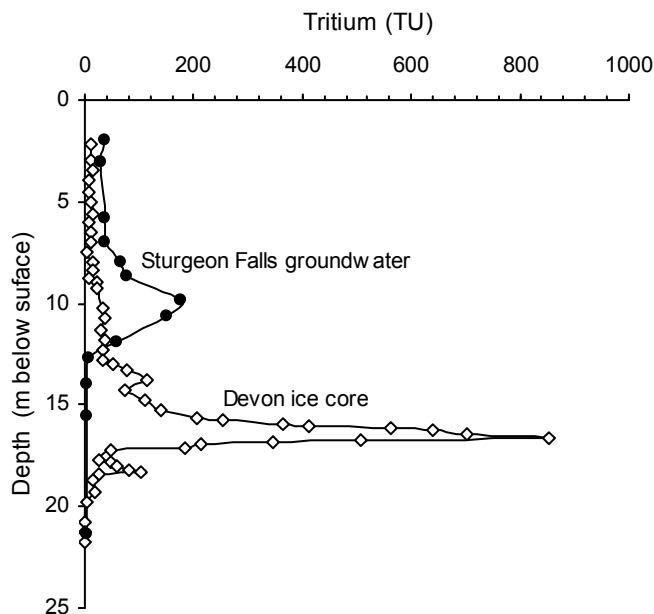


Fig. 9-5 Preservation of the 1963 bomb peak in precipitation measured in an ice core from Devon Island Ice Cap, and in downward moving groundwater in a sandy aquifer near Sturgeon Falls, Ontario. Note the highly attenuated peak in the groundwater due to hydrodynamic dispersion and decay, whereas the glacier ice experiences only decay.

Groundwater age from radioactive decay: Calculating groundwater age from decay of a known input concentration is valid only if atmospheric levels have been constant (i.e. recharge since a return to natural levels in the 1990s), and there has been no significant hydrodynamic dispersion (i.e. plug flow). The calculation uses the decay equation where the measured tritium concentration ${}^3\text{H}_t$ after some time, t , is equal to the initial concentration, ${}^3\text{H}_0$, times the exponential decay function:

$${}^3\text{H}_t = {}^3\text{H}_0 e^{-\lambda t}$$

From above, the decay term, $\lambda = \ln 2/t_{1/2}$. Using tritium's half-life, $t_{1/2} = 12.32$ years, this equation can be rewritten:

$$t = -17.77 \ln \frac{{}^3\text{H}_t}{{}^3\text{H}_0}$$

This equation is visually represented by the decay curves in Fig. 9-4. As the initial tritium concentration is not readily known, graphs such as this can be used to back-extrapolate to the input function to determine age.

Plug flow, with no hydrodynamic dispersion, is a condition that is seldom met in most aquifers. Nonetheless, in modern systems for which the input function is stable and known such calculations may give a first-order approximation of age. An improvement on this method uses multi-year recharge scenarios where the annual tritium concentrations are weighted by a distribution of ages such as shown in Fig. 9-1. The 5, 10 and 20 year scenarios in Fig. 9-4 represent revised input functions that account for hydrodynamic mixing along the flow path which results in a distribution of ages, each with its own original ${}^3\text{H}_0$ concentration.

Tritium-helium dating of groundwater: The requirement of knowing the tritium input function for calculating groundwater ages from tritium decay is eliminated if the parent-daughter decay equation is used. Measuring ${}^3\text{He}$, the product of tritium decay, under some circumstances, can allow much more precise ages to be calculated. The difficulty is that measuring helium isotopes is not routinely available and as it is done in only a few laboratories.

The measured concentration of tritium, ${}^3\text{He}_t$, can give the time, t , of decay (age) as long as the initial concentration ${}^3\text{H}_0$ is known:

$$^3\text{H}_t = ^3\text{H}_0 e^{-\lambda t}$$

However, given the complicated input function for tritium in precipitation (Fig. 9-2) knowing the initial value is not possible in most cases. On the other hand, the decay of tritium leads to an ingrowth of ^3He , which can be calculated from:

$$^3\text{He}_t = ^3\text{H}_0(1 - e^{-\lambda t})$$

By rearranging the tritium decay equation and substituting into the ^3He ingrowth equation, the input concentration of tritium $^3\text{H}_0$ is no longer a factor and now groundwater age, t , can be calculated from the measured tritium and ^3He concentrations:

$$^3\text{He}_t = ^3\text{H}_t(e^{\lambda t} - 1)$$

The measured helium concentration, $^3\text{He}_t$, is expressed in TU ($1\ ^3\text{He}$ per 10^{18} H). It must also be corrected for atmospheric ^3He that is dissolved at the time of recharge. Atmospheric He has a dissolved concentration that ranges from $5.0 \cdot 10^{-8}$ at 0°C to $4.6 \cdot 10^{-8}$ at 25°C and has a $^3\text{He}/^4\text{He}$ ratio in atmospherically-equilibrated water of $1.360 \cdot 10^{-6}$. This ratio is slightly less than the atmospheric ratio of $1.384 \cdot 10^{-6}$ due to fractionation during dissolution, which favours ^4He in water.

The measured value of ^3He , corrected for atmospheric ^3He , represents ^3He ingrown from ^3H decay, and is then used in the dating equation:

$$t = 17.77 \cdot \ln(1 + [^3\text{He}_t]/[^3\text{H}_t])$$

where $[^3\text{He}_t]/[^3\text{H}_t]$ is the concentration ratio of these two isotopes expressed in tritium units.

None-routine tools for dating modern groundwater

A number of isotopes methods can be used to assess mean residence times. Seasonal variations in stable isotopes provide a measure of age. The more routinely applied techniques are based on the decay of radionuclides (Table 1-3). Those with a long half-life (^{14}C , ^{36}Cl , ^{39}Ar and ^{81}Kr) can be used to date

paleo-groundwaters, and are discussed in Chapter 8. Short-lived radioisotopes (^3H , ^{32}Si , ^{37}Ar , ^{85}Kr and ^{222}Rn) and those produced by man's nuclear activities over the past four decades (^3H , ^{14}C , ^{36}Cl and ^{85}Kr) indicate modern recharge.

While the methods for analysis of ^3H and ^{14}C are now routine, other radioisotopes require complicated sampling and/or analytical techniques. The “submodern” period in the dating range between modern waters and paleo-groundwaters is problematic. While this >45 to ~1000 year range can potentially be filled by ^{39}Ar dating ($t_{1/2} = 256$ yr), this method requires rather ideal aquifers, very large samples, complicated sample preparation techniques and special counting facilities. Very few laboratories can afford this and, therefore, ^{39}Ar dating has not developed into a routine tool.

Thermonuclear ^{36}Cl

Thermonuclear bomb testing generated not only a whopping peak in tritium, but also in the radioactive isotope of Cl — ^{36}Cl . The high neutron flux of hydrogen bomb tests in marine settings during the early years of testing elevated concentrations by over two orders of magnitude above its natural atmospheric abundance. By consequence, ^{36}Cl can be used — like tritium — to identify modern recharge. However, unlike ^3H , its long half-life ($t_{1/2} \sim 300,000$ years) precludes the use of ^{36}Cl decay to date modern waters.

Natural atmospheric production of ^{36}Cl occurs due to activation of atmospheric argon by solar radiation and arrives at the earth's surface as a dry fallout or in precipitation. Natural production and fallout varies with latitude from close to $20 \text{ atoms m}^{-2} \text{ s}^{-1}$ at mid latitudes to less than 5 near the equator and poles. It is also produced epigenetically through activation of Cl, K and Ca on the land surface by solar radiation (see Chapter 8). Thermonuclear ^{36}Cl was produced by activation of atmospheric Ar and marine Cl^- by the high neutron fluxes accompanying low altitude tests in the 1950s. A record of this fallout is preserved in glacier ice, reconstructed by Bentley et al. (1986). High levels of ^{36}Cl in groundwater indicate, like tritium, that recharge has occurred since this time. **Error! Reference source not found.** shows that since the period of maximum ^{36}Cl contamination, concentrations have been steadily declining. This washing out of atmospheric Cl has brought concentrations, at least by 1980, back closer to natural levels. However, storage and recycling of Cl in the biosphere seems to maintain a background ^{36}Cl activity chronically elevated above natural levels (Gwen Milton, Atomic Energy of Canada Ltd., pers. comm.).

Concentrations of ^{36}Cl in groundwater requires conversion from the fallout rate (atoms $\text{m}^{-2} \text{s}^{-1}$) to concentration in water (atoms per litre). For example, with a natural ^{36}Cl fallout rate of $15 \text{ atoms m}^{-2} \text{s}^{-1}$, annual precipitation of 750 mm, and 75 mm loss to evapotranspiration, the concentration in runoff and/or groundwater can be calculated. The annual fallout rate would be:

$$15 \text{ atoms m}^{-2} \text{s}^{-1} = 473 \cdot 10^6 \text{ atoms m}^{-2} \text{yr}^{-1}$$

Dividing by the corrected annual precipitation gives 1:

$$473 \cdot 10^6 / (0.75 - 0.0075) = 700 \cdot 10^6 \text{ atoms m}^{-3} \text{ or } 7 \cdot 10^5 \text{ atoms L}^{-1}$$

Measurements of ^{36}Cl concentrations in groundwaters exceeding about 10^7 atoms L^{-1} (depending on rainfall and infiltration rates) is a good indication that there is a component of thermonuclear ^{36}Cl and hence, modern recharge. Remember that, unlike ^3H , ^{36}Cl is a solute only and so how and when the chloride was dissolved is important. Conjunctive use of ^{36}Cl and ^3H is of use in recharge studies (Chapter 4). Applications of ^{36}Cl as a dating tool for old groundwaters are found in Chapter 8.

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Questions

1. Account for the attenuated peak in the groundwaters from Sturgeon Falls, in Fig. 9-5. What is the average vertical velocity for this groundwater? Given a porosity of 0.3 and an annual precipitation of

1000 mm, what is the rate of recharge (mm/yr) and how much is lost to evapotranspiration.

2. What would be the age of a groundwater moving by plug flow that was recharged by precipitation with a tritium concentration of 8.4 TU?
3. A groundwater sample from a shallow sandy aquifer in the Ottawa area has a measured tritium concentration of 16 TU in 2004. Estimate its mean tritium age by simple decay with no dispersive mixing and using a 20 year mixing scenario (Fig. 9-4). What is the realistic range of average ages for this sample?

This groundwater was also sampled for helium concentration and isotopes, yielding values of $4.67\text{E-}08$ and $2.85\text{E-}06$ respectively. Determine the concentration of tritogenic ^3He in units of TU, and see if you can refine your age estimate.